Ferromagnetic 0– π Josephson junctions

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Abstract We present a study on low- T_c superconductorinsulator-ferromagnet-superconductor (SIFS) Josephson junctions. SIFS junctions have gained considerable interest in recent years because they show a number of interesting properties for future classical and quantum computing devices. We optimized the fabrication process of these junctions to achieve a homogeneous current transport, ending up with high-quality samples. Depending on the thickness of the ferromagnetic layer and on temperature, the SIFS junctions are in the ground state with a phase drop either 0 or π . By using a ferromagnetic layer with variable step-like thickness along the junction, we obtained a so-called $0-\pi$ Josephson junction, in which 0 and π ground states compete with each other. At a certain temperature the 0 and π parts of the junction are perfectly symmetric, i.e. the absolute critical current densities are equal. In this case the degenerate ground state corresponds to a vortex of supercurrent circulating clock- or counterclockwise and creating a magnetic flux which carries a fraction of the magnetic flux quantum

1 Introduction

Superconductivity (S) and ferromagnetism (F) are two competing phenomena. On one hand a bulk superconductor expels the magnetic field (Meissner effect). On the other hand the magnetic field for $H>H_{c2}$ destroys the superconductivity. This fact is due to the unequal symmetry in time: ferromagnetic order breaks the time-reversal symmetry, whereas conventional superconductivity relies on the pairing of time-reversed states. It turns out that the combination of both, superconductor and ferromagnet, leads to rich and interesting physics. One particular example – the phase oscillations of the superconducting Ginzburg-Landau order parameter inside the ferromagnet – will play a major role for the devices discussed in this work.

The current-phase relation $I_s(\phi)$ of a conventional SIS Josephson junction (JJ) is given by $I_s(\phi) = I_c \sin(\phi)$. $\phi = \theta_1 - \theta_2$ is the phase difference of the macroscopic superconducting wave functions $\Psi_{1,2} = \sqrt{n_s}e^{i\theta_{1,2}}$ (orderparameters of each electrode) across the junction, I_c is the critical current. Usually I_c is positive and the minimum of the Josephson energy $U = E_J(1 - \cos \phi)$, $E_J = \frac{I_c \Phi_0}{2\pi}$ is at $\phi = 0$. However, Bulaevskii et al. [1] calculated the supercurrent through a JJ with ferromagnetic impurities in the tunnel barrier and predicted a negative supercurrent, $I_c < 0$. For $-I_c \sin(\phi) = 0$ the solution $\phi = 0$ is unstable and corresponds to the maximum energy $U = E_J(1 + \cos \phi)$, while $\phi = \pi$ is stable and corresponds to the ground state. Such JJs with $\phi = \pi$ in ground state are called π junctions, in contrast to conventional 0 junctions with $\phi = 0$. In case of a π Josephson junction the first Josephson relation is modified to $I_s(\phi) = -I_c \sin(\phi) = I_c \sin(\phi + \pi)$. In experiment the measured critical current in a single junction is always positive and is equal to $|I_c|$. It is not possible to distinguish 0 JJs from π JJs from the current-voltage characteristic (IVC) of a single junction. The particular $I_c(T)$ [2] and $I_c(d_F)$ [3] dependencies for SFS/SIFS type junction are used to determine the π coupled state. For low-transparency SIFS junctions the $I_c(d_F)$ dependence is given by

$$I_c(d_F) \propto \exp\left(\frac{-d_F}{\xi_{F1}}\right) \cos\left(\frac{d_F - d_F^{\text{dead}}}{\xi_{F2}}\right),$$
 (1)

where ξ_{F1}, ξ_{F2} are the decay and oscillation lengths of critical current and $d_F^{\rm dead}$ is the dead magnetic layer thickness [4]. For $\frac{1}{2}\xi_{F2}\pi < d_F - d_F^{\rm dead} < \frac{3}{2}\xi_{F2}\pi$ the coupling in ground state of JJs is shifted by π .

In a second work Bulaevskii et al. [5] predicted the appearance of a spontaneous supercurrent at the boundary between a 0 and a π coupled long JJ (LJJ). This supercurrent emerges in the absence of a driving bias current or an external field H, i.e. in the ground state. Depending on the length of the junction L the supercurrent carries one half of the flux quantum, i.e. $\Phi_0/2$ (called

M. Weides et al.

semifluxon), or less. Fig. 1(a) depicts the cross section of a symmetric $0-\pi$ long JJ. The spontaneous supercurrent j_s flows either clockwise or counterclockwise, creating the magnetic field of $\pm \Phi_0/2$. The current density jumps from maximum positive to maximum negative value at the $0-\pi$ phase boundary. A theoretical analysis based on the perturbed sine-Gordon equation is given in Ref. [6]. Below we will first discuss the properties of the spontaneous supercurrent and, second, various systems having $0-\pi$ phase boundaries.

2

Spontaneous supercurrent Kirtley et al. [7] calculated the free energy of $0-\pi$ JJs for various lengths of the 0 and π parts as a function of the normalized length $\ell = L/\lambda_J$ and the degree of asymmetry $\Delta = |j_c^{\pi}| L_{\pi}/|j_c^0| L_0$, where j_c^0, j_c^{π} are the critical current densities and L_0, L_{π} are the lengths of 0 and π parts respectively, so that $L = L_0 + L_{\pi}$. The state of a symmetric $0-\pi$ junction $(\Delta = 1)$ with spontaneous flux has lower energy than the states $\phi = 0$ or $\phi = \pi$ without flux. Symmetric $0-\pi$ junctions have

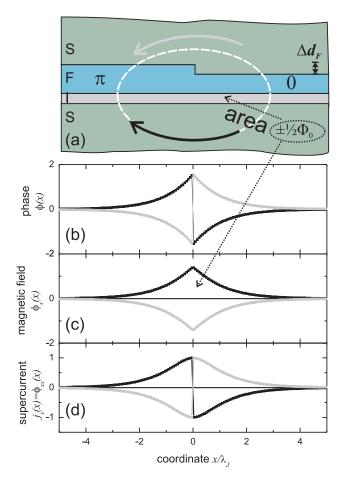


Fig. 1 (a) Sketch of a $0-\pi$ SIFS JJ with step-like thickness of F-layer and circulating supercurrent j_s around $0-\pi$ phase boundary. The junction length $L\gg \lambda_J$, therefore the spontaneous flux (area below magnetic field) is equal to half of a flux quantum Φ_0 (semifluxon). (b)-(d) depicts the phase $\phi(x)$, magnetic field $\phi_x(x)$ and supercurrent $j_s(x) = \frac{I_c}{|I_c|} \sin \phi$ of the $0-\pi$ junction.

always some self-generated spontaneous flux, although its amplitude vanishes for $L \to 0$ as $\Phi \approx \Phi_0 \ell^2/8\pi$. For example, a symmetric $0-\pi$ JJ of the total length $L = \lambda_J$ has a spontaneous magnetic flux $\Phi \approx 0.04\Phi_0$ and a symmetric $0-\pi$ JJ with $L = 8\lambda_J$ has a spontaneous flux of some 2-3% below $\Phi_0/2$. Only in case of a infinitely long JJ we refer to the spontaneous flux as semifluxons, for shorter JJs it is named fractional vortex.

The supercurrent or magnetic flux can be directly detected by measuring $I_c(H)$ [7], by scanning SQUID (superconducting quantum interference device) microscopy (in the LJJ limit, see [8, 9]) or by LTSEM (low temperature scanning electron microscopy) [10].

 $0-\pi$ junctions technology $0-\pi$ Josephson junctions with a spontaneous flux in the ground state are realized with various technologies. The presence of fractional vortex has been demonstrated experimentally in d-wave superconductor based ramp zigzag junctions [9], in long Josephson $0-\pi$ junctions fabricated using the conventional Nb/-Al-Al₂O₃/Nb technology with a pair of current injectors [11], in the so-called tricrystal grain-boundary LJJs [8, 12, 13] or in SFS/SIFS JJs [14, 15, 16] with stepped ferromagnetic barrier as in Fig. 1. In the latter systems the Josephson phase in the ground state is set to 0 or π by choosing proper F-layer thicknesses d_1 , d_2 for 0 and π parts, i.e. the amplitude of the critical current densities j_c^0 and j_c^{π} can be controlled to some degree. The advantages of this system are that it can be prepared in a multilayer geometry (allowing topological flexibility) and it can be easily combined with the well-developed Nb/Al-Al₂O₃/Nb technology.

The starting point for estimation of the ground state of a stepped JJ is studying the IVCs and $I_c(H)$ for the planar reference 0 and π JJs. From this one can calculate important parameters such as the critical current densities j_c^0, j_c^{π} , the Josephson penetration depths $\lambda_J^0, \lambda_J^{\pi}$ and the ratio of asymmetry Δ . For $0-\pi$ junctions one needs 0 and π coupling in *one* junction, setting high demands on the fabrication process. The ideal $0-\pi$ JJ would have equal $|j_c^0| = |j_c^{\pi}|$ and a $0-\pi$ phase boundary in its center to have a symmetric situation. Furthermore the junctions should be underdamped (SIFS structure) since low dissipation is necessary to study dynamics and eventually macroscopic quantum effects. The junctions should have a high j_c (and hence small $\lambda_J \propto \sqrt{j_c}$) to reach the LJJ limit and to keep high $V_c = I_c R$ products, where V_c is the characteristic voltage and R the normal state resistance.

Previous experimental works on $0-\pi$ JJs based on SFS technology [14, 15] gave no information about j_c^0 and j_c^{π} . Hence, the Josephson penetration depth λ_J could not be calculated for these samples and the ratio of asymmetry Δ was unknown. The first intentionally made symmetric $0-\pi$ tunnel JJ of SIFS type with a large V_c was realized by the authors [16], making direct transport measure-

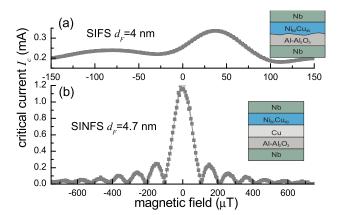


Fig. 2 (Color online) $I_c(H)$ of (a) SIFS (4 nm NiCu) and (b) SINFS (Cu 2 nm, NiCu 4.7 nm) stacks. Oxygen pressure is 0.45 mbar for SIFS and 0.015 mbar for SINFS type.

ments of $I_c(H)$ and calculation of the ground state with spontaneous flux feasible.

Within this paper we review the physics of $0-\pi$ coupled SIFS-type Josephson junctions and give an overview on our experimental results. Special focus is put on the fabrication of SIFS junctions having a planar or stepped-typed ferromagnetic layer (NiCu), the determination of ground state (0 or π for planar JJs) and asymmetry of critical currents (stepped JJs). Finally we give an estimation of the spontaneous magnetic flux in the ferromagnetic $0-\pi$ JJs.

2 Fabrication

The fabrication process for planar junctions is based on Nb/Al-Al₂O₃/NiCu/Nb stacks, deposited by dc magnetron sputtering [17]. Thermally oxidized 4-inch Si wafer served as substrate. First of all, a 120 nm thick Nb bottom electrode and a 5 nm thick Al layer were deposited. Second, the aluminium was oxidized for 30 min at room temperature in a separate chamber. Third, the ferromagnet (i.e. $Ni_{60}Cu_{40}$ alloy, $T_C = 225$ K) was deposited. To have many structures with different thicknesses in one fabrication run, we decided to deposit a wedge-shaped F-layer. For this the substrate and sputter target were shifted about half of the substrate diameter. This allowed the preparation of SIFS junctions with a gradient in F-layer thickness in order to minimize inevitable run-to-run variations. The sputtering rates for NiCu along the gradient were determined by thickness measurements on reference samples using a Dektak profiler. At the end a 40 nm Nb cap layer was deposited. The tunnel junctions were patterned using a three level optical photolithographic mask procedure and Ar ionbeam milling [18]. The insulation between top and bottom electrode is done by a self-aligned growth of Nb₂O₅ insulator by anodic oxidation of Nb after the ion-beam etching. The Nb₂O₅ exhibited a defect free insulation between the superconducting electrodes.

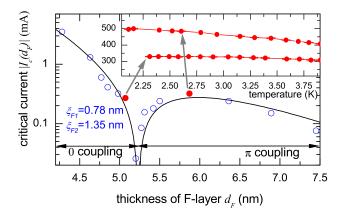


Fig. 3 (Color online) $I_c(d_F)$ and $I_c(T)$ (inset) dependences of SIFS junctions at 4.2 K. Note the difference of the slope of $I_c(T)$ for 0 and π coupled junction (inset).

Topological and electrical measurements, see Ref. [17], indicated that the direct deposition of NiCu on the tunnel barrier (SIFS-stacks) led to an anomalous $I_c(H)$ dependence such as shown in Fig. 2(a), which is an indication for an inhomogeneous current transport. An additional 2 nm thin Cu layer between the Al_2O_3 tunnel barrier and the ferromagnetic NiCu (SINFS-stacks) brought considerable benefits, as it ensured a homogeneous current transport, see Fig. 2(b). In this way a high number of functioning devices with j_c spreads less than 2% was obtained. The variation of the F-layer thickness over a length of one junction diameter is less than 0.02 nm. For simplification we refer in the following to SIFS stacks, although the actual multilayer is SINFS-type.

The patterning of stepped junctions was done after the complete deposition of the planar SIFS stack and before the definition of the junction mesa by argon-etching and Nb₂O₅ insulation. The detailed process is published in Ref. [19]. The junction was partly protected with photoresist to define the step location in the F-layer, followed by i) selective reactive etching of the Nb, ii) ionetching of the NiCu by Δd_F and iii) subsequent in situ deposition of Nb. To our knowledge, this was the first controlled patterning of $0-\pi$ JJs based on a ferromagnetic interlayer.

The planar 0, π reference junctions and the stepped $0-\pi$ junctions were fabricated from a single trilayer.

3 SIFS junctions without step-like F-layer

All investigated junctions had an area of $10~000\,\mu\text{m}^2$, but the length and width were different for different junctions. The length was comparable or shorter than the Josephson penetration depth λ_J . We investigated the thickness dependence of the critical current $I_c(d_F)$. To produce the Al₂O₃ barrier the Al layer was oxidized at 0.015 mbar yielding $j_c \approx 4.0~\text{kA/cm}^2$ for the reference superconductor-insulator-superconductor (SIS) JJs. Then SIFS stacks with wedge-like F-layer were fabricated in

M. Weides et al.

another run. Taking the JJs of the same geometry ($100 \times 100 \ \mu\text{m}^2$), but situated at different places on the wafer (i.e. different d_F) we have measured the nonmonotonic $I_c(d_F)$ dependence shown in Fig. 3. As a result the fitted parameters are $\xi_{F1} = 0.78$ nm, $\xi_{F2} = 1.35$ nm and $d_F^{\text{dead}} \approx 3.09$ nm. The coupling changed from 0 to π at the crossover thickness $d_F^{0-\pi} = \frac{\pi}{2}\xi_{F2} + d_F^{\text{dead}} = 5.21$ nm [4].

The magnetic and spin-orbit scattering in the F-layer mixes the up and down spin states of electrons in the conduction bands. If the spin-flip scattering time τ_s is short $\hbar \tau_s^{-1} \gg k_B T_c$, like in NiCu alloys, the temperature dependence of scattering provides the dominant mechanism for the $I_c(T)$ dependence [20]. The oscillation period ξ_{F2} becomes shorter for decreasing temperature, thus the whole $I_c(d_F)$ dependence is squeezed to thinner F-layer thicknesses. Hence, the temperature dependence of the critical current $I_c(T)$ is an interplay between an increasing component due to an increasing gap and a magnetic coupling dependent contribution which may de- or increase I_c . The $I_c(T)$ relations for two JJs (one 0, one π) are shown in the inset of Fig. 3. At $d_F = 5.11$ nm the JJ is 0 coupled, but one can relate the nearly constant I_c below 3.5 K to the interplay between an increasing gap and a decreasing oscillation length $\xi_{F2}(T)$. The $d_F = 5.87$ nm JJ is π coupled and showed a linearly increasing I_c with decreasing temperature.

4 SIFS junctions with step-like F-layer

Various structures on the wafer were placed within a narrow ribbon perpendicular to the gradient in the Flayer thickness and were replicated along this gradient. One ribbon contained reference JJs with the uniform F-layer thickness d_1 (uniformly etched) and d_2 (nonetched) and a JJ with a step Δd_F in the F-layer thickness from d_1 to d_2 . The lengths L_{d_1} and L_{d_2} are both equal to 167 μ m. The lithographic accuracy is of the order of 1 μ m. A set of structures with difference in d_F between neighboring ribbons of 0.05 nm was obtained. Comparing the critical currents I_c of non-etched JJs (dots), see Fig. 4 with the experimental $I_c(d_F)$ data for the etched samples (stars) we estimate the etchedaway F-layer thickness as $\Delta d_F \approx 0.3$ nm. The stars in Fig. 4 are shown already shifted by this amount. Now we choose the set of junctions which have the thickness d_2 and critical current $I_c(d_2) < 0$ (π junction) before etching and have the thickness $d_1 = d_2 - \Delta d_F$ and critical current $I_c(d_1) \approx -I_c(d_2)$ (0 junction) after etching. One option is to choose the junction set denoted by closed circles around the data points in Fig. 4, i.e. $d_1 = 5.05 \text{ nm}$ and $d_2 = 5.33$ nm.

The I-V characteristics and the magnetic field dependence of the critical current $I_c(H)$ was measured for all three junctions: 0 JJ with $d_F=d_1,\,\pi$ JJ with $d_F=d_2$ and $0\text{-}\pi$ JJs with stepped F-layer (d_1 and d_2

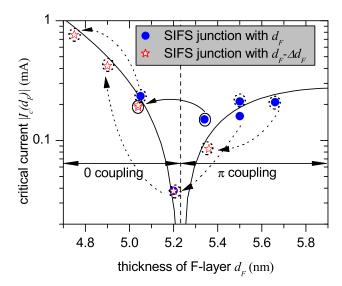


Fig. 4 (Color online) Critical current I_c of the uniformly etched (star) and non-etched (dot) SIFS junctions versus the F-layer thickness before etching d_F . The fit of the experimental data for non-etched samples using Eq.(1) is shown by the continuous line. The JJs were oxidized at 0.015 mbar.

in each half). The magnetic diffraction pattern $I_c(H)$ of the $0-\pi$ JJ and the 0 and π reference JJs are plotted in Fig. 5. The magnetic field H was applied in-plane of the sample and parallel to the step in the F-layer. Due to a small net magnetization of the F-layers the $I_c(H)$ of references junctions were slightly shifted along the Haxis. Nevertheless, both had the same oscillation period $\mu_0 H_{c1} \approx 36 \ \mu \text{T}$. At $T \approx 4.0 \ \text{K}$ the $0-\pi$ JJs was slightly asymmetric with $I_c^0 \approx 208 \ \mu \text{A}$ and $I_c^{\pi} \approx 171 \ \mu \text{A}$ (data of reference JJs). To achieve a more symmetric configuration, the bath temperature was reduced, because a decrease in temperature should increase $I_c^{\pi} = I_c(d_2)$ more than $I_c^0 = I_c(d_1)$, like for the 0 and π samples in the inset of Fig. 3. As a result, both $I_c^0(T)$ and $I_c^{\pi}(T)$ were increasing when decreasing the temperature, but with different rates. At $T \approx 2.65$ K the critical currents I_c^0 and I_c^{π} became approximately equal, see Fig. 5. The magnetic field dependence of the planar reference junctions $I_c^0(H)$ and $I_c^{\pi}(H)$ look like perfect Fraunhofer patterns. One can see that the $I_c^0(H)$ and $I_c^{\pi}(H)$ measurements almost coincide, having the form of a symmetric Fraunhofer pattern with the critical currents $I_c^0 \approx 220 \ \mu\text{A}, I_c^{\pi} \approx 217 \ \mu\text{A}$ and the same oscillation period. The stepped $0-\pi$ junction had a magnetic field dependence $I_c^{0-\pi}(H)$ with a clear minimum near zero field and almost no asymmetry. The critical currents at the left and right maxima (146 μA and 141 μA) differ by less than 4 %, i.e. the $0-\pi$ junction is symmetric, and its ground state in absence of a driving bias or magnetic field (I = H = 0)can be calculated [16]. Our symmetric $0-\pi$ LJJ had an normalized length of $\ell = 1.3$, with a spontaneous flux in the ground state of

$$\pm \Phi \approx \Phi_0 \ell^2 / 8\pi \approx 0.067 \cdot \Phi_0$$

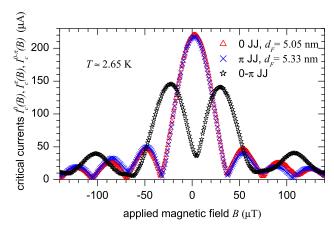


Fig. 5 (Color online) $I_c(H)$ of $0-\pi$ JJ (open triangles) with H applied parallel to short axis, overlayed with the nonetched (dot) and etched (stars) reference SIFS junction measurements. At $T\approx 2.65$ K the $0-\pi$ JJ becomes symmetric. The junction dimensions are $330\times 30~\mu\mathrm{m}^2$.

being equal to 13% of $\Phi_0/2$. A detailed calculation taking several deviations from the ideal short JJ model into account can be found elsewhere [21].

5 Summary

The concept and realization of 0- π junction based on SIFS stacks has been presented. The realization of π coupling in SIFS junctions and the precise combination of 0 and π coupled parts in a single junction has been shown. The coupling of the ferromagnetic Josephson tunnel junctions was investigated by means of transport measurements. The emergence of a spontaneous flux, which was calculated as 13% of half a flux quantum $\Phi_0/2$, was observed in the magnetic field dependence of the current-voltage characteristics of the 0- π JJ.

As an outlook, the ferromagnetic $0-\pi$ Josephson junctions allow to study the physics of fractional vortices with a good temperature control of the symmetry between 0 and π parts. We note that symmetry is only needed for JJ lengths $L \lesssim \lambda_J$. For longer JJs the semifluxon appears even in rather asymmetric JJs, and T can be varied in a wide range affecting the semifluxon properties only weakly. The presented SIFS technology allows us to construct 0, π and $0-\pi$ JJs with comparable j_c^0 and j_c^π in a single fabrication run. Such JJs may be used to construct classical and quantum devices such as oscillators, memory cells, π flux qubits [22, 23] or semifluxon based qubits [24].

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